

# Nature of superconducting state in the new phase in (TMTSF)<sub>2</sub>PF<sub>6</sub> under pressure.

L. P. Gor'kov and P. D. Grigoriev\*

*National High Magnetic Field laboratory, Florida State University, Tallahassee, Florida*

(Dated: February 6, 2008)

The unusual phase has been recently observed in the organic material (TMTSF)<sub>2</sub>PF<sub>6</sub>, where superconductivity (SC) coexists with spin-density wave (SDW) in the pressure interval  $p_{c1} < p < p_c$  below the first order transition into SC or normal metal phase. Assuming that the coexistence takes place on the microscopic scale, we consider the properties of the intermediate phase. We show that the new superconducting state inside SDW phase just above  $p_{c1}$  must bear a triplet pairing.

PACS numbers: 71.30.+h, 74.70.Kn, 75.30.Fv

Below the critical pressure,  $p_c$ , destroying spin-density wave (SDW) in the quasi-one-dimensional (Q1D) organic compound (TMTSF)<sub>2</sub>PF<sub>6</sub> a pressure interval  $p_{c1} < p < p_c$  has been discovered,[1, 2] in which the dielectric SDW and metallic/SC regions coexist spatially. The details of this coexistence are not entirely determined experimentally; in particular, the domain sizes of the coexisting phases remain unknown. While in [1] a macroscopic size was assumed for domains, the reason for macroscopic coexistence at fixed pressure is unclear yet. Instead of this, the spatially inhomogeneous phase, called the soliton phase (SP), has been assumed[3] in the pressure region  $p_{c1} < p < p_c$ . The emergent SP is then ascribed to the appearance of metallic domain walls above  $p_{c1}$ . This phenomenon has been first proposed for the charge-density waves.[4] The experimental data on NMR[5] and on AMRO[6] about the domain size do not contradict the assumption of Ref. [3].

One of the most interesting questions in this context is the question about the origin and properties of the SC in this new state. As it was shown recently,[7] superconductivity appears first at  $p_{c1}$ ; at higher pressure  $T_c^{SC}$  increases and reaches the value of SC transition temperature in the metallic state. The mechanism and the type of SC in the normal phase (above the critical pressure,  $p_c$ , for the first order phase transition) still remains unknown, though some arguments in favor of the triplet pairing have been suggested.[8] In the new intermediate state ( $p_{c1} < p < p_c$ ) the absence of the Knight shift change[9] and too high critical field  $H_{c2}$  compared to the values of critical temperature[2, 10] attract special attention. In the present letter we address the issue of the type of SC pairing in the intermediate phase.

Although in Ref. [3] the onset of SP was suggested at  $p > p_{c1}$ , the alternative destructive mechanism of the gapped SDW state could be realized as a gradual formation of electron-hole ungapped pockets when pressure enhances the "antinetesting" term of the quasi-1D electronic spectrum in (TMTSF)<sub>2</sub>PF<sub>6</sub> (for CDW such a mechanism was discussed in [11, 12]). It turns out that close to  $p_{c1}$ :

$p - p_{c1} \ll p_c$ , the SC onset can be studied analytically for the two scenarios: of weakly overlapping solitons in SP or at the appearance of small ungapped e-h pockets on the background of the homogeneous SDW. The main result below is that close to  $p_{c1}$  in both scenarios the low-temperature Cooper instability exists only for the triplet pairing.

The quasi-1D compound (TMTSF)<sub>2</sub>PF<sub>6</sub> in normal state is characterized by the two open Fermi surface (FS) sheets with the spectrum

$$\varepsilon(\mathbf{k}) = v_F(|k_x| - k_F) + t_\perp(\mathbf{k}_\perp). \quad (1)$$

In the SC state the Gor'kov order parameter at each (left or right) FS has the form

$$\begin{aligned} f_{\alpha\beta}^{LR}(\mathbf{r}) &= \langle \hat{\Psi}_\alpha^L(\mathbf{r}) \hat{\Psi}_\beta^R(\mathbf{r}) \rangle; \\ f_{\alpha\beta}^{RL}(\mathbf{r}) &= \langle \hat{\Psi}_\alpha^R(\mathbf{r}) \hat{\Psi}_\beta^L(\mathbf{r}) \rangle. \end{aligned} \quad (2)$$

The spatial inversion symmetry in (TMTSF)<sub>2</sub>PF<sub>6</sub> allows to classify the pairing type by the symmetry of the order parameters in Eq. (2):  $f_{\alpha\beta}^{LR} = \pm f_{\alpha\beta}^{RL}$ , where the sign ( $\pm$ ) depends on whether the SC pairing has singlet (+) or triplet (-) character. For simplicity, we use the mean-field model, in which only the backward scattering matrix element,  $g_1$ , between electrons on the opposite sheets contributes to the SC pairing.[13] The convenience of such a model is that in the metallic phase the Cooper instability would always manifest itself at some  $T_c^{SC}$  for triplet or singlet pairing depending on the sign of the coupling constant  $g_1$  in the familiar relation[14, 15]

$$1 = [g_1 \ln(\bar{\omega}/T_c^{SC})]^2, \quad (3)$$

where  $\bar{\omega}$  is a proper cutoff, and  $g_1$  is the matrix element of the backward scattering interaction multiplied by the density of states at the Fermi level.

Before to apply the Cooper instability analysis to the phase with the SDW, one needs first to determine the wave functions and the energy spectrum of the latter. To achieve this goal we generalized the approach developed for CDW[11] to the SDW case. In particular, this approach allows the treatment of the homogeneous SDW and of the SP on equal footing. As shown in Ref. [11], the spectrum of Eq. (1) allows the exact mapping of

\*Also at L. D. Landau Institute for Theoretical Physics, Chernogolovka, Russia; Electronic address: pashag@itp.ac.ru

the anisotropic Q1D problem onto the purely 1D one, where sophisticated methods for studying solitons have been developed.[16] As in Ref. [11], we consider the general case of SDW order parameter,  $\hat{\Delta}_{SDW}(\mathbf{r})$ , acquiring spatial modulation in the presence of the soliton walls:

$$\hat{\Delta}_{SDW}(\mathbf{r}) = \Delta_{SDW}(x) \cos(\mathbf{Q}\mathbf{r})(\vec{\sigma}\mathbf{1}). \quad (4)$$

The Shroedinger equation writes  $\hat{H}_{\mathbf{k}}^0 \Psi_{\mathbf{k}} = \varepsilon_{\mathbf{k}} \Psi_{\mathbf{k}}$  with the Hamiltonian

$$\hat{H}_{\mathbf{k}}^0 = \begin{pmatrix} \hat{\varepsilon}(\mathbf{k}_{\perp}) - \frac{iv_F d}{dx}; & \Delta_{SDW}(x)(\vec{\sigma}\mathbf{1}) \\ \Delta_{SDW}^*(x)(\vec{\sigma}\mathbf{1}); & \hat{\varepsilon}(\mathbf{k}_{\perp} - \mathbf{Q}_{\perp}) + \frac{iv_F d}{dx} \end{pmatrix} \quad (5)$$

and with the four-component (spin) wave function

$$\Psi_{\mathbf{k}} \equiv \begin{pmatrix} \psi_{\mathbf{k}}^R(x) \\ \psi_{\mathbf{k}-\mathbf{Q}}^L(x) \end{pmatrix}, \quad \psi_{\mathbf{k}}^{R(L)}(x) = \begin{pmatrix} \psi_{\mathbf{k}\uparrow}^{R(L)}(x) \\ \psi_{\mathbf{k}\downarrow}^{R(L)}(x) \end{pmatrix}, \quad (6)$$

which combines the electron wave functions on the right and left Fermi surface sheets, denoted by  $R(L)$  superscripts. Transformation

$$\begin{aligned} \psi_{\mathbf{k}_{\perp}\alpha}^R(x) &= \exp\{ix[k_x - \varepsilon_{-}(\mathbf{k}_{\perp})/v_F]\} \psi_{\alpha}^R(x) \\ \psi_{\mathbf{k}_{\perp}-\mathbf{Q}_{\perp}\alpha}^L(x) &= \exp\{-ix[k_x - \varepsilon_{-}(\mathbf{k}_{\perp})/v_F]\} \psi_{\alpha}^L(x), \end{aligned} \quad (7)$$

where

$$\varepsilon_{\pm}(\mathbf{k}_{\perp}) = [t(\mathbf{k}_{\perp}) \pm t(\mathbf{k}_{\perp} - \mathbf{Q}_{\perp})]/2, \quad (8)$$

reduces the Hamiltonian (5) to

$$\hat{H}_{1D} = \begin{pmatrix} -iv_F d/dx & \Delta(x)(\vec{\sigma}\mathbf{1}) \\ \Delta^*(x)(\vec{\sigma}\mathbf{1}) & iv_F d/dx \end{pmatrix}. \quad (9)$$

The eigenvalues of the 3D problem (5) are

$$\varepsilon_{\lambda, \mathbf{k}_{\perp}} = E_{\lambda} + \varepsilon_{+}(\mathbf{k}_{\perp}), \quad (10)$$

where the index  $\lambda$  numerates the eigenvalues of the 1D Hamiltonian (9) for a periodic soliton lattice  $\Delta_{SDW}(x)$ . Finding  $\Delta_{SDW}(x)$  is a separate problem, that can be solved exactly for the commensurate case[17] and in the limit of a single soliton.[18] For homogeneous SDW  $\Delta_{SDW}(x) = \text{const}$  the analysis of Eqs. (5),(6) can be easily performed in the momentum representation. The quasiparticle energy spectrum (10) than becomes

$$\varepsilon_{1,2}(\mathbf{k}) \equiv \varepsilon^{+}(\mathbf{k}_{\perp}) \pm \sqrt{\xi^2 + |\Delta_{SDW}|^2}, \quad (11)$$

where  $\xi \equiv v_F(|k_x| - k_F) - \varepsilon^{-}(\mathbf{k}_{\perp})$ .

The idea behind the calculation in both cases is that at  $p > p_{c1}$  a branch of the energy spectrum crosses the chemical potential. For a network of the rarefied soliton walls, a single soliton wall may be treated as metallic sheets[3, 4] with the thickness  $d \sim \xi_0 = \hbar v_F / T_{SDW}$ . At higher pressure solitons will overlap and form a 3D metallic band that lies inside the SDW gap. For the pockets' scenario at  $p > p_{c1}$ , the transverse dispersion  $\varepsilon_{+}(\mathbf{k}_{\perp})$  in

Eq. (10) becomes greater than the SDW energy gap, forming first open electron-hole pockets of the form

$$\varepsilon(\mathbf{k}) = \pm \delta \pm [a_1 (\Delta \mathbf{k}_{\perp})^2 + b_1 \xi^2], \quad (12)$$

where

$$\begin{aligned} \delta &\equiv |\Delta_{SDW} - t_{\perp}(\mathbf{k}_0)| \ll \Delta_{SDW}, \\ a_1 &\sim t'_{\perp} b^2 \text{ and } b_1 \approx 1/2 \Delta_{SDW}. \end{aligned}$$

In each case the formed small "Fermi surface" is subject to the examination for a possible Cooper instability. Such an instability, should it occurs at some low temperature, would signify the possibility for onset of SC pairing.

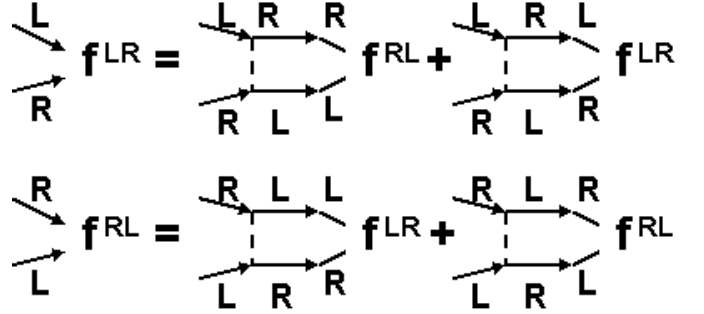


FIG. 1: The diagram equations for the functions  $f_{\alpha\beta}^{LR}$  and  $f_{\alpha\beta}^{RL}$  in the presence of SDW. The solid lines represent the electron Green functions:  $G_{i\omega_n}^{R(L)R(L)}$ . The dash lines represent the short-range interaction (in our case the backward scattering) of electrons.

Analysis of the Cooper ladder diagrams with the interaction  $g_1$  in Eq. (3) can be carried out using the standard methods for searching logarithmically divergent terms at  $T \rightarrow 0$ . [15] In our case calculations are more tedious being complicated by the underlying SDW structure. We briefly sketch the main steps in the calculations. The corresponding diagram equations are shown in Fig. 1. The lines in the Cooper bubble stand for the proper Greens functions. Compared to pairing in the normal state, there are additional terms that come from "non-diagonal" Greens functions,  $G^{RL}$  and  $G^{LR}$ , when two Fermi surface sheets of Eq. (1) mix together in the SDW(CDW) presence. The  $4 \times 4$  (spin) matrix Greens functions can be presented in the general form

$$\hat{G}_{i\omega_n}(\mathbf{r}, \mathbf{r}') = - \sum_{\lambda, \mathbf{k}_{\perp}} \frac{\Psi_{\mathbf{k}}^{\dagger}(\mathbf{r}') \otimes \Psi_{\mathbf{k}}(\mathbf{r})}{i\omega_n - \varepsilon_{\lambda, \mathbf{k}_{\perp}}}, \quad (13)$$

where  $\Psi_{\mathbf{k}}(\mathbf{r})$  is given by Eq. (6).

Calculations are transparent for a homogeneous SDW(CDW). We discuss the case in which small e-h pockets (12) get formed at some point  $\mathbf{k}_0$  on the FS. In the momentum space the Green functions entering the Cooper block in Fig. 1 write down as

$$g^{RR(LL)}(\mathbf{k}, \omega) = \frac{i\omega - \varepsilon(\mathbf{k})}{[i\omega - \varepsilon_1(\mathbf{k})][i\omega - \varepsilon_2(\mathbf{k})]} \quad (14)$$

where  $\omega \equiv \omega_n = (2n+1)\pi T$ ,  $\varepsilon(\mathbf{k})$  is given by Eq. (1) and  $\varepsilon_{1,2}$  are given by Eq. (11). The nondiagonal Green functions  $\hat{g}^{LR(RL)}(\mathbf{k}, \omega) = (\vec{\sigma}\vec{l})g^{LR(RL)}(\mathbf{k}, \omega)$ , with

$$g^{LR}(\mathbf{k}, \omega) = \frac{\Delta_{SDW}}{[i\omega - \varepsilon_1(\mathbf{k})][i\omega - \varepsilon_2(\mathbf{k})]} \quad (15)$$

and  $g^{RL}(\mathbf{k}, \omega) = [g^{LR}(\mathbf{k}, -\omega)]^*$ .

The sum of two Cooper bubbles in the right-hand part of equations, shown schematically in Fig. 1, writes down as

$$\begin{aligned} \hat{f}^{LR} = & -Tg_1 \sum_{\mathbf{k}, \omega} \left[ g^{RR}(\mathbf{k}, \omega) \hat{f}^{RL} g^{LL}(-\mathbf{k}, -\omega) \right. \\ & \left. + (\vec{\sigma}\vec{l})g^{LR}(\mathbf{k}, \omega) \hat{f}^{LR} (\vec{\sigma}\vec{l})^T g^{RL}(-\mathbf{k}, -\omega) \right]. \end{aligned} \quad (16)$$

The ultraviolet logarithmic divergence in (16) comes only from the first line; it would give the term  $|g_1| \ln(\bar{\omega}/C\Delta_{SDW})$ , where  $C = \text{const} \sim 1$ . Once electrons and hole pockets open at the Fermi level, they lead to the appearance of the low-energy divergence that contribute an additional logarithmic divergent term  $\sim \ln(\sqrt{\delta\Delta_{SDW}}/T)$ , where  $\delta$  determines the size of small pockets. The total equation (16) for the SC transition temperature than rewrites as

$$\ln(C\Delta_{SDW}/T_{SC}) = A \ln(\sqrt{\delta\Delta_{SDW}}/T). \quad (17)$$

(Remember that  $\Delta_{SDW} \approx 12K \gg T_{SC} \approx 1K$ .) Calculations of this contribution from the single pocket go as follows. Consider Eq. (12) for one pocket. The energy level  $\varepsilon_1(\mathbf{k})$  crosses the chemical potential at the point  $\mathbf{k} = \mathbf{k}_0$  to form a small FS at pressure slightly above  $p_{c1}$ . Then  $\varepsilon_2(\mathbf{k}) \approx 2\Delta_{SDW}$  at  $\mathbf{k}$  near  $\mathbf{k}_0$ . Similarly, taking in the nominator of Greens functions  $\varepsilon(\mathbf{k}) \approx \Delta_{SDW}$  at  $\mathbf{k}$  near  $\mathbf{k}_0$  and substituting the simplified Greens functions (14),(15) into (16) we find a familiar form of the logarithmic divergence at low temperature coming from the poles of the Greens functions:

$$\sum_{\mathbf{k}, \omega} \frac{T}{\omega^2 + \varepsilon_1^2(\mathbf{k})} = \sum_{\mathbf{k}} \frac{\tanh[\varepsilon_1(\mathbf{k})/2T]}{2\varepsilon_1(\mathbf{k})}.$$

Substituting (12) for  $\varepsilon_1(\mathbf{k})$  and changing the variables  $a(\Delta\mathbf{k}_\perp)^2 \rightarrow y^2$  and  $b(k_x - k_F)^2 \rightarrow x^2$  we obtain the integral of the form

$$\begin{aligned} & \frac{1}{\sqrt{a_1 b_1}} \int \frac{\tanh[\varepsilon_1(\mathbf{x}, \mathbf{y})/2T]}{2\varepsilon_1(\mathbf{x}, \mathbf{y})} \frac{dxdy}{(2\pi)^2} \\ = & \frac{1}{\sqrt{a_1 b_1}} \int_0^\delta \frac{\tanh[(\delta - r^2)/2T]}{\delta - r^2} \frac{dr^2}{8\pi} \\ & + \frac{1}{\sqrt{a_1 b_1}} \int_0^{\Delta_{SDW}} \frac{\tanh[(\delta - r^2)/2T]}{\delta - r^2} \frac{dr^2}{8\pi} \\ \sim & \frac{\ln[\sqrt{\Delta_{SDW}\delta}/T]}{4\pi}. \end{aligned}$$

Returning to Eq. (17), the value of the prefactor  $A$ , which defines the SC transition temperature, is just a number. Most remarkable, however, is the observation that  $A$  drastically depends on the type of pairing. For spin-singlet pairing the spin structure of the SC order parameter  $\hat{f}^{LR} = \hat{f}^{LR} = i\hat{\sigma}_y f^{LR}$ , and using  $\hat{\sigma}_y(\vec{\sigma}\vec{l})^T = -(\vec{\sigma}\vec{l})\hat{\sigma}_y$ , one rewrites equation (16) as

$$\begin{aligned} 1 = & -Tg \sum_{\mathbf{k}, \omega} [g^{RR}(\mathbf{k}, \omega) g^{LL}(-\mathbf{k}, -\omega) \\ & - g^{LR}(\mathbf{k}, \omega) g^{RL}(-\mathbf{k}, -\omega)]. \end{aligned} \quad (18)$$

The second line in this equation acquires the sign “-” due to the spin structure of the background SDW phase, which is in contrast to the SC on the CDW background. This difference in the sign leads to the cancelation in the main approximation of the low-energy logarithmic singularity in (18) for the chosen pocket at  $\mathbf{k} = \mathbf{k}_0$ . This results in a smallness of the factor  $A \sim \delta/\Delta_{SDW}$  before the logarithm in the r.h.s. of Eq. (16).

This cancelation may not occur for triplet pairing. Substituting the spin structure of triplet order parameter,  $\hat{f}^{LR} = (\hat{\sigma}\vec{\mathbf{d}})\hat{\sigma}_y f^{LR}$ , together with  $f^{RL} = -f^{LR}$  into (16) and using  $(\vec{\sigma}\vec{l})(\hat{\sigma}\vec{\mathbf{d}})\hat{\sigma}_y(\vec{\sigma}\vec{l})^T = (\hat{\sigma}\vec{\mathbf{d}})\hat{\sigma}_y - 2(\vec{\mathbf{d}}\vec{l})(\vec{\sigma}\vec{l})\hat{\sigma}_y$  we obtain in the right hand part of Eq. (16)

$$\begin{aligned} T \sum_{\mathbf{k}, \omega} & \left[ -g^{RR}(\mathbf{k}, \omega) (\hat{\sigma}\vec{\mathbf{d}}) g^{LL}(-\mathbf{k}, -\omega) \right. \\ & \left. + g^{LR}(\mathbf{k}, \omega) g^{RL}(-\mathbf{k}, -\omega) \left\{ (\hat{\sigma}\vec{\mathbf{d}}) - 2(\vec{\mathbf{d}}\vec{l})(\vec{\sigma}\vec{l}) \right\} \right]. \end{aligned} \quad (19)$$

We see that the main infrared divergent terms cancel each other only if  $\vec{\mathbf{d}} \perp \vec{l}$ . For  $\vec{\mathbf{d}} \parallel \vec{l}$  the factor  $A$  is the same, as in the case of the CDW background.

Analysis for the onset of SC in the soliton wall scenario goes through in the similar fashion. The logarithmic singularity of the Cooper type via the isolated soliton wall sheets has already been discussed for CDW.[12] For SDW one has to return to Eq. (16) and the wave functions (6) making use of the exact single soliton solution[18]. One can easily check that similar cancelation in the nominator depending on the spin structure happens in this scenario also.

To summarize, we have shown that at either way the SDW is being destroyed by pressure above  $p_{c1}$ , SC in this new state is expected to bear triplet character. This result also shows the remarkable difference between SDW and CDW coexisting with superconductivity on a single conducting band. Our results, although have been derived assuming  $|p - p_{c1}| \ll p_{c1}$ , should extend over a considerable part of the new phase in (TMTSF)<sub>2</sub>PF<sub>6</sub> at  $p_{c1} < p < p_c$  if there is no additional phase transition at  $p < p_c$ .

The work was supported by NHMFL through the NSF Cooperative agreement No. DMR-0084173 and the State

of Florida, and (PG) in part, by DOE Grant DE-FG03-03NA00066 and RFBR N 06-02-16551.

- 
- [1] T. Vuletic, P. Auban-Senzier, C. Pasquier et al., Eur. Phys. J. B **25**, 319 (2002).
  - [2] I. J. Lee, M. J. Naughton, G. M. Danner, and P. M. Chaikin, Phys. Rev. Lett. **78**, 3555 (1997); I. J. Lee, P. M. Chaikin, and M. J. Naughton, Phys. Rev. B **62**, R14 669 (2000); I. J. Lee, P. M. Chaikin, and M. J. Naughton, Phys. Rev. Lett. **88**, 207002 (2002).
  - [3] L.P. Gor'kov, P.D. Grigorev, Europhys. Lett. **71**, 425 (2005).
  - [4] S.A. Brazovskii, L.P. Gor'kov, J.R. Schrieffer, Physica Scripta **25**, 423 (1982).
  - [5] S.E. Brown, 2005 (unpublished).
  - [6] I. J. Lee, S. E. Brown, W. Yu, M. J. Naughton, and P. M. Chaikin, Phys. Rev. Lett. **94**, 197001 (2005).
  - [7] The Orsay group, private communication, 2006 (unpublished).
  - [8] L. P. Gor'kov and D. Jerome, J. Phys. (Paris) Lett. **46**, L-643 (1985).
  - [9] I. J. Lee, S. E. Brown, W. G. Clark, M. J. Strouse, M. J. Naughton, W. Kang, and P. M. Chaikin, Phys. Rev. Lett. **88**, 017004 (2002); I.J. Lee, D. S. Chow, W. G. Clark, M. J. Strouse, M. J. Naughton, P. M. Chaikin, and S. E. Brown, Phys. Rev. B **68**, 092510 (2003).
  - [10] I. J. Lee, P. M. Chaikin, and M. J. Naughton, Phys. Rev. B **65**, 180502(R) (2002).
  - [11] S.A. Brazovskii, L.P. Gor'kov, A.G. Lebed', Sov. Phys. JETP **56**, 683 (1982) [Zh. Eksp. Teor. Fiz. **83**, 1198 (1982)].
  - [12] L.P. Gor'kov, A.G. Lebed', Journal de Physique **44**, C3-1531 (1983).
  - [13] We have checked that our result does not change in the general case, when the forward scattering is also taken into account.
  - [14] H. Suhl, B.T. Matthias, and L.R. Walker, Phys. Rev. Lett. **3**, 552 (1959)
  - [15] A.A. Abrikosov, L.P. Gor'kov and I.E. Dzyaloshinskii, "Methods of quantum field theory in statistical physics", Dover Publications, INC., New York 1977.
  - [16] S.A. Brazovskii and N.N. Kirova, Sov. Sci. Rev. A Phys., **5**, 99 (1984).
  - [17] S.A. Brazovskii, S.A. Gordyunin and N.N. Kirova, JETP Lett. **31**, 451 (1980)[Pis'ma v ZhETF **31**, 486 (1980)].
  - [18] S.A. Brazovskij, Sov. Phys. JETP **51**, 342 (1980).